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Application of Aliquat-336 nitrate ionic liquid based extractants for minor actinide separation

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The presence of minor actinides in spent nuclear fuel poses a major obstacle for both open and closed fuel cycles. Several long-lived isotopes of Am and Cm are responsible for the main heat production and high radiotoxicity of the nuclear waste. These isotopes make both reprocessing as well as final disposal complicated and costly. In the framework of the Belgian MYRRHA-project, a Gen IV prototype accelerator-driven fast reactor is foreseen to be built, in which these long-lived minor actinide isotopes can be efficiently burnt.¹

If we consider a closed fuel cycle (and thus reprocessing), U and Pu are separated from spent fuel via the so-called PUREX process, leaving a PUREX-raffinate containing the fission products and minor actinides. In order to separate a small amount of trivalent actinide elements from the PUREX-raffinate, several sophisticated methods have been elaborated (TALSPEAK, DIAMEX, SANEX or GANEX, etc.), all of which are based on the use of aliphatic diluents such as dodecane or kerosene. These diluents however all have common shortcomings, namely they are volatile, flammable and sensitive to radiation-induced degradation that reduces their process lifetime. By the application of a non-volatile and non-flammable and radiation-resistant diluent medium for the minor actinide separation, secondary waste production can be reduced.

[Aliquat-336][NO₃] and benzyl-trioctylammonium nitrate ionic liquids as diluents and TODGA as extractant ligand was used for the liquid-liquid extraction of lanthanides and actinides from nitric acid solutions. Batch extractions using stable lanthanide isotopes together with tracers of ¹⁵²Eu, ²⁴¹Am and ²⁴⁴Cm isotopes were conducted to determine the distribution ratios, separation factors, and kinetics of the extraction, and also the loading effect on the phase behavior of the ionic liquids. The actinide and lanthanide ions are extracted via a neutral solvation mechanism and fast kinetics with TODGA. From a slope analysis of log $D_{Am(III)}$ vs. log [TODGA] plot the number of extracting molecules participating in the metal-ligand complex was determined.

The possibility of directly separating the chemically similar trivalent actinides from trivalent lanthanides was also studied using actinide-selective soft-donor BTBP ligands.

These initial experiments shall be followed by radiation stability studies on the ionic liquid to simulate realistic extraction circumstances. Degradation effects will be analysed by NMR and ESI-MS studies and in addition, the effect of radiation on the extraction system will be evaluated (effect on distribution ratios and separation factors).

¹ <http://myrrha.sckcen.be/>